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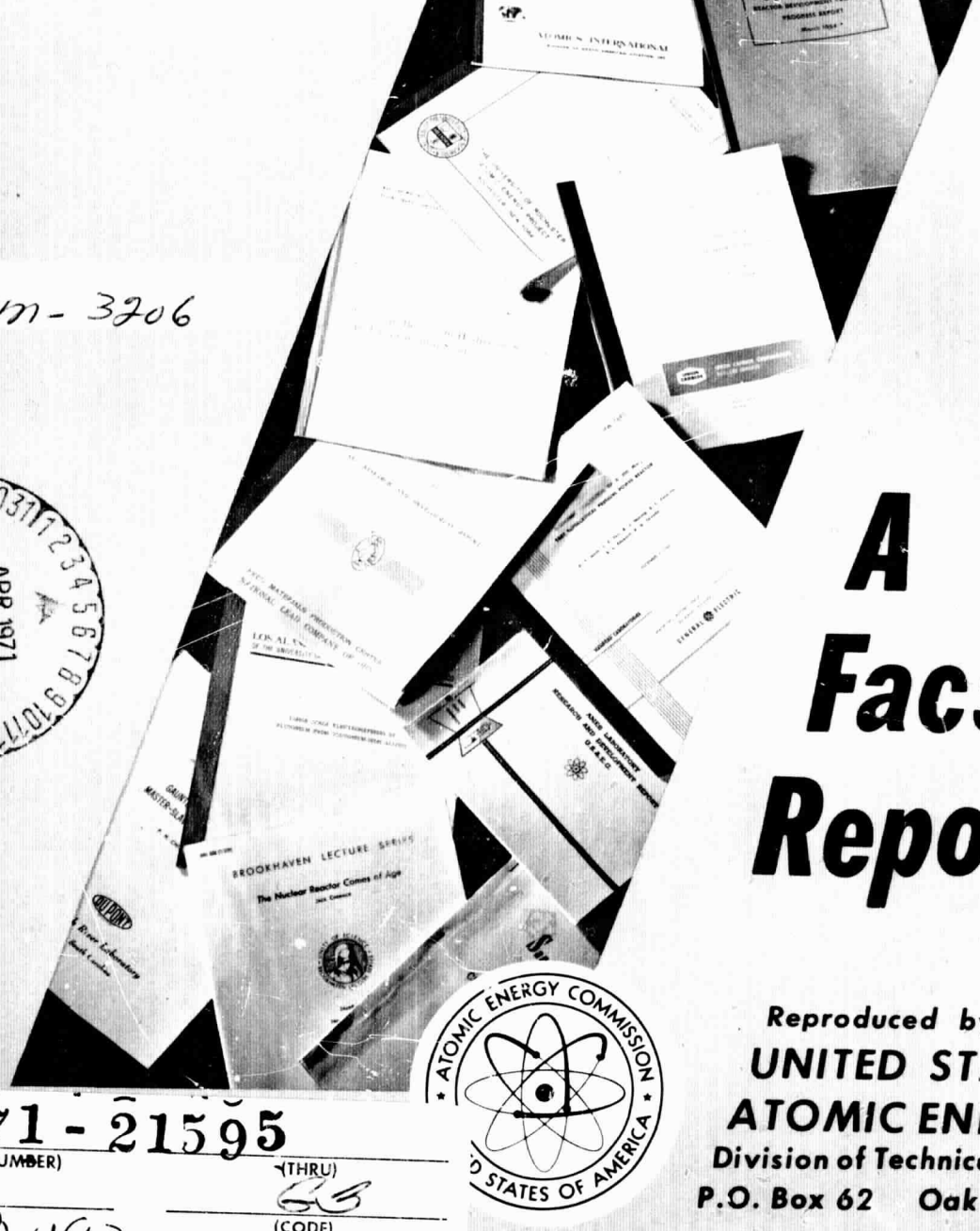

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COMPARISON OF CALCULATED AND MEASURED LOWER CRITICAL FIELD FOR SOME Nb-Ti ALLOYS

M. S. Lubell and R H. Kernohan

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COMPARISON OF CALCULATED AND MEASURED LOWER CRITICAL FIELD FOR SOME Nb-Ti ALLOYS*

M. S. Lubell and R. H. Kernohan
Oak Ridge National Laboratory†
Oak Ridge, Tennessee 37830

ABSTRACT

An expression is obtained from the Maki theory for $H_{c1}(0)$ in terms of three measurable material constants T_c, ρ_n, and $H^*_{c2}(0)$. The calculations are compared to the measured H_{c1} vs T extrapolated to T = 0 K for three vacuum annealed Nb-Ti alloys. Values of $H_{c1}(0)$ are also determined from the computation of Harden and Arp. Both sets of calculated values differ greatly from the measured values showing that the Maki theory is not applicable for $\kappa_{GL} < 30$, and the computation of Harden and Arp is not valid at T = 0 K in disagreement with recent results of Echarri et al. Analysis of data available in the literature supports our conclusions. Comparison of the magnetization data near T_c with the theory of Neumann and Teworit shows reasonable agreement.

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1. INTRODUCTION

Calculations of the lower critical field H_{c1} in units of the thermodynamic critical field H_c are not presently available for the complete range of reduced temperature and Ginzburg-Landau parameter κ_{GL} [1]. It would be useful in ac applications to be able to calculate H_{c1} from easily obtainable material constants and to avoid the necessity of magnetization measurements.

Harden and Arp [2] have numerically solved an equation derived from the Ginzburg-Landau theory and extended Abrikosov's calculation of H_{c1}/H_c for all $\kappa_{GL} > 1/\sqrt{2}$. However, this result is generally believed to be valid (as was Abrikosov's original solution) only at temperatures close to T_c. Maki [3] was the first to obtain a solution for the temperature dependence of H_{c1} for dirty superconductors ($\xi_o \gg \ell$) but only in the limit of large κ_{GL}. The range of validity of Maki's theory (hereafter M) has not been adequately tested by experiments partly owing to the difficulty in measuring H_{c1} in materials with a large κ_{GL} and small electron mean-free-path ℓ. The thermodynamic critical field is also difficult to measure unambiguously in hysteretic superconductors.

Recently Echarri *et al.* [4] (hereafter E) have shown for a Mo-34% Re alloy with a $\kappa_{GL} = 5.8$ that the calculations of H_{c1} using Harden and Arp (hereafter HA) gave excellent agreement with their measurements of H_{c1} at a reduced temperature of $t = 0.356$. This agreement would seem to indicate that the HA computation might be valid over a larger temperature range than originally anticipated. Their measurements of H_{c1} at 4.2 K ($t = 0.356$) are also compared with the M prediction for H_{c1} at T = 0 K and do not seem to warrant their conclusion that the M theory is restricted

to $\kappa > 50$. Since there is no well established temperature function for H_{c1}, a zero temperature value could not be obtained from a 4.2 K measurement. If, however, a quadratic temperature variation for $H_{c1}(T)$ is assumed, then calculations based on the M theory yield values that on the average are only 7% lower than the extrapolated measured ones. Since as they state, the temperature variation of H_{c1} is slower than parabolic, then the difference between the M theory and the experimental values is even less than this, which is quite surprising.

We have investigated a series of Nb-Ti alloys covering a range of κ_{GL} values from 5 to 24 in which all the material constants necessary for a calculation of $H_{c1}(0)$ have been individually measured. These were compared with experimental values of $H_{c1}(0)$ determined by a smooth extrapolation of H_{c1} vs T data to T = 0 K. In this manner, it was seen whether the M theory would be applicable to lower κ_{GL} than originally anticipated. Also the HA calculation, valid for all κ_{GL} at $T = T_c$, was independently checked for its validity at T = 0 K even though agreement could hardly be expected. Magnetization data taken near T_c was in reasonable agreement with theory especially when mean-free-path corrections were considered.

2. DERIVATION OF FORMULA USED IN ANALYSIS

Starting with the M theory as our point of departure and employing well-known type II relationships, a simple formula for $H_{c1}(0)$ is obtained below in terms of easily measurable parameters. Maki [3] gave for $H_{c1}(0)$, in the dirty limit ($\xi_o \gg \ell$),

$$\frac{H_{c1}(0)}{H_c(0)} = \frac{\ln \kappa_3(0)}{\sqrt{2}\,\kappa_3(0)} \qquad (1)$$

where $\kappa_3(0) = 1.272\ \kappa_1(0) = 1.53\ \kappa_{GL}$ is one of the three generalized Ginzburg-Landau parameters which are all equal to κ_{GL} at $T = T_c$. Equation (1) was independently derived by Melik-Barkhudarov [5] who obtained $\kappa_3(0) = 1.54\ \kappa_{GL}$ and $\kappa_3(1) = \kappa_{GL}$. The upper critical field at zero temperature is given by [1] $H_{c2}(0) = \sqrt{2}\ \kappa_1(0)\ H_c(0)$. Substituting these expressions in Eq. (1), we find

$$H_{c1}(0) = \frac{H_c^2(0)}{1.272\ H_{c2}(0)} \ln\left(1.272\ H_{c2}(0)/\sqrt{2}\ H_c(0)\right). \qquad (2)$$

An expression for the calculation of the thermodynamic critical field at zero temperature in terms of measurable constants is readily obtainable. Combining a BCS equation [1] $H_c(0) = 2.43\ \gamma^{1/2}\ T_c$ and a formula due to Kim et al. [6] $H^*_{c2}(0) = 3.11 \times 10^{-5}\ \gamma\rho_n T_c$, we obtain for $H_c(0)$ in G,

$$H_c(0) = 436\left(H^*_{c2}(0)\ T_c/\rho_n\right)^{1/2}, \qquad (3)$$

where $H^*_{c2}(0)$ is the nonparamagnetically limited WHAG upper critical field at zero temperature in kG, T_c is the transition temperature for zero field in Kelvins, ρ_n is the normal state resistivity in $\mu\Omega$-cm, and γ is the normal state electronic specific heat coefficient in ergs/cm^3-K^2. The Kim et al. [6] expression for $H^*_{c2}(0)$ assumes $\kappa_{GL} = \kappa_\ell$ and thus is valid only in the dirty limit [7] which however is a good approximation for this alloy system which has a low intrinsic κ_o. Substituting Eq. (3) into Eq. (2) gives the desired result for $H_{c1}(0)$ in G,

$$H_{c1}(0) = \left(74.6\ T_c/\rho_n\right) \ln\left(4.26\ H^*_{c2}(0)\ \rho_n/T_c\right). \qquad (4)$$

3. EXPERIMENTAL

As a test of the range of applicability of Eq. (4), measurements of T_c, ρ_n, $H^*_{c2}(0)$, and $H_{c1}(T)$ were made on three Nb-Ti alloys of nominal composition Nb-5% Ti, Nb-10% Ti, and Nb-25% Ti (atomic %).

The samples were made from an ingot which was arc-cast on a copper hearth in an argon atmosphere from starting materials 99.9% pure. After a slight amount of cold working, the ingot underwent a homogenization anneal at 1250°C for 2 h and was fast-quenched in ice water. The ingot was drawn down to 0.77 mm diam wire which was measured and then vacuum annealed at 1400°C for 2 h. Further details on sample preparation have been given [8].

In Table I the experimentally determined quantities for the three alloys are tabulated. The normal resistivity ρ_n was measured by a standard four probe dc technique at T = 4.2 K in a field higher than the upper critical field. The upper critical field at zero temperature was determined by a flux flow experiment from the Kim et al. [6] expression $\rho_f/\rho_n = H/H^*_{c2}(0)$ where the flux flow resistance $\rho_f = dV/dI$ is given by the slope of the linear portion of the voltage vs current curve taken at constant H and T_B. An example of flux flow data for the annealed Nb-25% Ti sample is shown in Fig. 1. The critical temperature T_c and lower critical field $H_{c1}(T)$ were measured magnetically on a bundle of six 2.6 cm lengths of wire each 0.77 mm in diameter. The critical temperature was taken as the midpoint of a transition having a spread of only 20 mK, however the absolute accuracy was no better than $\pm$ 50 mK. The error in the determination of ρ_n and $H^*_{c2}(0)$ is not known exactly, but it is less than 10%. Values of H_{c1} were obtained by measuring small remanent magnetic moments

after increasingly higher excursions of the applied field into the Meissner region (perfect diamagnetic state where B = 0). Extrapolation of the remanent moment vs applied field to zero moment then yielded values of H_{c1}. Although the sensitivity of the magnetization equipment [9] is sufficient to detect a flux density of 1 G, there is still some scatter in the data shown in Fig. 2, possibly because of the geometry (wire bundle). This method generally yields values less than those obtained by a determination of the first point of the departure from linearity of the Meissner region. The value of H_{c1} at T = 0 K was obtained by a smooth extrapolation of the data as shown in Fig. 2.

The critical temperature was about 0.1 K lower in the annealed samples than in the as drawn cold-worked material. Both H^*_{c2} and ρ_n were on the order of 10% lower in the annealed material than in the cold-worked samples. Lower values of H_{c1} than shown in Table I could possibly have been obtained by either a higher temperature anneal or a longer anneal, but the accompanying loss of Ti would have had a deleterious effect on the distribution of Ti in each sample.

4. CALCULATIONS

The values calculated from the measurements are shown in Table II. The thermodynamic critical field $H_c(0)$ was calculated from Eq. (3). Kappa one at T = 0 K $\kappa_1(0)$ was determined from the measured $H^*_{c2}(0)$ and the calculated $H_c(0)$ which is equivalent to taking $\kappa_1(0) = 1.203\ \kappa_{GL}$ where κ_{GL} is assumed equal to κ_ℓ, the extrinsic value only. Hence $\kappa_1(0)$ is also calculated from the three measured quantities ρ_n, T_c, and $H^*_{c2}(0)$ since by this procedure $\kappa_1(0) = 1.62\left(H^*_{c2}(0)\ \rho_n/T_c\right)^{1/2}$. Kappa three at T = 0 K $\kappa_3(0)$ was calculated from $1.272\ \kappa_1(0)$.

The first column of $H_{c1}(0)$ values listed in Table II was determined from the graph of HA using $\kappa_3(0)$ and $H_c(0)$ calculated from Eq. (3). As is readily evident, these values are very much below the measured ones. The calculation of $H_{c1}(0)$ using Eq. (4) derived from the M theory is given in the last column. The disagreement in this case is even worse. These calculations average between 36% and 57% below the measured values. The disagreement is particularly bad for the sample with higher Ti content, i.e. larger κ_{GL}.

5. DISCUSSION

The discrepancy between the calculated and measured values of $H_{c1}(0)$ is due to either grossly inaccurate measurements, poor assumptions in utilizing the theories of the calculated values, or inapplicability of the theories. Let us examine each of these possibilities in turn.

Of the measured parameters, $H^*_{c2}(0)$ is in agreement with values reported in the literature [10, 11] for other compositions in the high Nb concentration end of the Nb-Ti system. Our T_c measurements do not agree with Fietz and Webb [10] (hereafter FW) who indicate only 0.1 K change from Nb to Nb-12.5% Ti while our values increase monotonically. We have checked the Vickers hardness for our three compositions, and in qualitative agreement with the T_c data it also shows a monotonic increase with increasing Ti content varying from 78 to 96 to 123 diamond pyramid hardness for the 5, 10, and 25% Ti cold-worked alloys, respectively. The measuring thermometer is unlikely to have a significant systematic error since the T_c of pure Nb measured with it was 9.26 K which is in agreement with many recently reported measurements. The normal resistivity is the

easiest and most straightforward parameter to measure, and the only source of difficulty leading to an error is the measurement of the exact length between voltage probes. The measurements of $H_{c1}(T)$ on a hysteretic material such as Nb-Ti are difficult. As a criterion for determining H_{c1}, we have used the first detectable remanent moment equivalent to detecting a flux of 10^{-1} G-cm^2. This procedure gives a value that is even less than that obtained by trying to determine the exact deviation from linearity of an M vs H graph, i.e. the end of the Meissner state. The only value in the literature to compare our results to is a measurement on Nb-3% Ti by DeSorbo [12]. He gives $H_{c1}(4.2\ K) \approx 675$ G which should be and is larger than $H_{c1}(4.2\ K) = 520$ G for our 5% Ti alloy. One would, perhaps, anticipate an anomalously high value of H_{c1} due to surface effects as first proposed by Bean and Livingston [13]. Our specimens did not have smooth surfaces which tend to enhance the formation of an image barrier [14] but instead were left in the roughened state characteristic of the drawing process.

Since there is no simple analytic form for the temperature dependence of H_{c1}, we can only say that the ratio $H_{c1}(T)/H_{c2}(T)$ does increase with decreasing T which is in accordance with the theory of Neumann and Teworit [15]. This result should be true regardless of the value of κ_{GL} or the ratio ξ_o/ℓ. The temperature dependence of $H_{c1}(T)$ was close but not equal to a quadratic one. A final check on the consistency of our data is given by the calculation of $H_c(0)$ from the three measured quantities $H^*_{c2}(0)$, ρ_n, and T_c utilizing Eq. (3). The values shown in Table II are between the calculated and measured values given by FW on annealed Nb-Ti alloys. The calculation of $H_c(0)$ brings up the first discrepancy between theory and experiment. FW have shown that the

calculated BCS $H_c(0)$ using an estimated rather than measured γ is on the average about 18% less than the measured $H_c(0)$ for annealed specimens for compositions covering the range from Nb to Nb-9% Ti. If the same is true for our samples and this is by no means at all certain, then this would account for some of the difference between our calculated and measured $H_{c1}(0)$. Although H_c could not be measured at low temperatures due to the relatively low applied field available, it was determined at a few high temperature points for the annealed samples, and the extrapolated value at T = 0 K using a quadratic temperature dependence yielded values within 5% of the calculated values shown in Table II.

In applying the theory we used forms containing $H^*_{c2}(0)$ [viz. Eqs. (3) and (4)], the nonparamagnetically limited upper critical field at zero temperature. However this is the value determined from flux flow experiments. In addition the GLAG limit should be dominant for Nb-Ti alloys in this range [16], and paramagnetic limiting probably is absent in all three compositions but most certainly in the 5 and 10% alloys. If the Ginzburg-Landau theory is to be applicable in the dirty limit below T_c, then κ_{GL} must be replaced by $\kappa_3(T)$. The procedure we have followed here in calculating $H_{c1}(0)$ is to replace $\kappa_3(0)$ in the M theory by $1.53\ \kappa_{GL}$ where κ_{GL} is given by the extrinsic component κ_ℓ. We have determined κ_ℓ through a calculation involving the normal state resistance. There are other methods [10, 17] for determining κ_{GL}, but these generally yield values within 10% of each other. Since $\kappa_{GL} \geq 5$ for all compositions, a variation of 10% in the magnitude of κ_{GL} would affect the final results by less than 5%. Finally there is one more check that can be made to substantiate the general validity of our calculations. Equation (1)

when combined with the definitions preceeding Eq. (2) in such a way as to eliminate $H_c(0)$ can be put in a form where $H_{c1}(0)$ is a function only of two parameters, i.e., $H_{c2}(0)$ and $\kappa_1(0)$.

$$H_{c1}(0) = \left(H_{c2}(0)/2.544\ \kappa_1^2(0)\right) \ell_n\ 1.272\ \kappa_1(0) \qquad (5)$$

By extrapolating the $H_{c2}(T)$ and $\kappa_1(T)$ data given by FW to T = 0 K, we have obtained values of $H_{c2}(0)$ and $\kappa_1(0)$ for their annealed Nb-4.5% Ti, Nb-9% Ti. The values of $H_{c1}(0)$ computed from Eq. (5) are 436 and 314 G for Nb with 4.5% and 9% Ti, respectively. This agreement between our values and those calculated from entirely different measurements (based on measurements of H_{c2} and H_c) lends further support to the conclusion that our measurements and methods of calculation are not seriously in error.

6. MAGNETIZATION MEASUREMENTS NEAR T_c

Although magnetization measurements yielding values of $H_{c2}(T)$ and $H_c(T)$ could only be performed at a couple of temperatures close to T_c, we present an analysis of this data for two reasons. First, we will show that an extrapolation of this data to low temperatures yields results close to our calculations. Second, a comparison of the data near T_c with theory applicable for this region shows reasonable agreement which provides confidence that the samples are not adversely contaminated with impurities.

A calculation of $H_{c1}(0)$ using our high temperature measurements of $H_c(T)$ and $\kappa_1(T) = H_{c2}(T)/\sqrt{2}\ H_c(T)$ extrapolated to T = 0 K using a quadratic

approximation for $H_c(T)$ and the M theory for $\kappa_1(T)$ gave results within 5% of those presented in Table II for the 5 and 10% samples. Only one high temperature evaluation was obtained for the 25% sample and so an extrapolation was not practical, but a crude estimate gave a 40% higher value for $H_{c1}(0)$ which nevertheless still did not agree well with the measurement. The high temperature values of $\kappa_1(T)$ determined magnetically are compared to the T = 0 K calculations based on electrical measurements in Fig. 4.

In Fig. 5 the measured values of $H_{c1}(T)/H_c(T)$ are compared with the theories of M, HA, and Neumann and Tewordt [15]. The M result was obtained from the theoretical value of $\kappa_3(T)/\kappa_1(T)$ in the region near T_c, the measure $\kappa_1(T)$, and Eq. (1). Note that unlike the zero temperature calculations given previously, here the discrepancy between the M theory and the measurements decreases as κ_{GL} increases (i.e. increasing Ti). Results based on the HA computation using $\kappa_3(T)$ are in closer agreement with experiment than the M theory. The calculations using the Neumann and Tewordt [15] computation includes a correction for mean-free-path dependence. In calculating the mean-free-path parameter, we used a method similar to FW in determining the intrinsic Ginzburg-Landau kappa, κ_c.

We attempted to use our measured values of $H_{c1}(T)$ and calculated values of $H_c(T)$ (using both a BCS and a quadratic temperature dependence) to obtain "observed values" of $\kappa_3(T)$ based on the M theory, Eq. (1) but over a good part of the temperature range, there was no solution.

7. CONCLUSION

The fact that the calculations of $H_{c1}(0)$ differ so greatly from the measurements leads us to conclude that the M theory is not applicable for

type II superconductors with Ginzburg-Landau kappa values less than 26. One might anticipate a smaller discrepancy as κ increases toward high values but such was not the case for our samples. We have also concluded that one is not justified in using the computations of HA for zero temperature evaluations. In order to make more comparisons, we have calculated $H_{c1}(0)$ for other low kappa systems, namely the Nb-Ta data of Ogasawara et al. [18], the Mo-15% Re and Mo-25% Re data of Joiner and Blaugher [19], the Mo-32.3% Re data of Lerner et al. [20], and the Nb-Ta data by Ikushima and Mizusaki [21]. For all of the above cases, $H_c(0)$ was measured and did not have to be calculated. Only a summary of the results are given in Table III. The widely different values for Mo-32.3% Re are perhaps due to the two-phase nature of the composition [22]. Similarly the agreement between theory and experiment by E on a composition close to that used by Lerner et al. [20] might be a fortuitous consequence of measurements on a multi-phase material. In all cases, the value of $H_{c1}(0)$ calculated from Eq. (4) was lower than the measured H_{c1} extrapolated to T = 0 K. For low κ_{GL} material, the HA calculation in almost all cases agreed more closely with experiment than the M theory. Nevertheless the agreement is not good enough to warrant the conclusion that the HA results are valid over a wide temperature range. It has also been pointed out recently by Decker and Lacquer [23] that the M theory does not agree with experiment even for a high kappa ($\kappa_{GL} \sim 60$) material. Extension of the GLAG theories covering temperatures far from T_c valid for all κ_{GL} are still needed. Even if obtained, however, agreement with experiment may well require additional refinements such as inclusion of anisotropy of the Fermi-surface, multiple band effects, and as suggested by E normal state spin effects on the lower critical field.

8. ACKNOWLEDGMENTS

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TABLE I

Measured Values of Annealed Nb-Ti Alloys

Nominal Composition (At. %)	T_c (K)	ρ_n ($\mu\Omega$-cm)	$H^*_{c2}(0)$ (kG)	$H_{c1}(0)$ (G)
Nb-5% Ti	9.41	6.42	18	675
Nb-10% Ti	9.61	12.1	35	500
Nb-25% Ti	9.93	35.5	90.5	350

TABLE II

Calculated Values of the Nb-Ti Alloys

Nominal Composition (At. %)	$H_c(0)$ (G) Eq. (3)	$\kappa_1(0)$ (a)	$\kappa_3(0)$ (b)	$H_{c1}(0)$ (G) (c)	$H_{c1}(0)$ (G) Eq. (4)
Nb-5% Ti	2240	5.7	7.2	556	433
Nb-10% Ti	2300	11	14	364	310
Nb-25% Ti	2200	29	37	174	151

[a] Determined from the measured value $H^*_{c2}(0)$ and calculated $H_c(0)$ which is equivalent to $\kappa_1(0) = 1.203\ \kappa_{GL} = 1.62 \left(H^*_{c2}(0)\ \rho_n/T_c\right)^{1/2}$.

[b] Calculated from $\kappa_3(0) = 1.272\ \kappa_1(0) = 1.53\ \kappa_4$.

[c] Determined from the graph of Harden and Arp [2], using $\kappa_3(0)$ and the calculated $H_c(0)$.

TABLE III

Percent Difference Between Calculated from Measured $H_{c1}(0)$ from Analysis of Published Data Where H_c was Measured

Material	Eq. (4)	Eq. (1)	HA
Nb-Ta[a]	- 37	- 47	- 26
Mo-Re[b]	- 35	- 37	- 11
Mo-32.3% Re[c]	- 50[e]	- 11	+ 20
Nb-Ta[d]	- 46	- 64	- 30

[a]Reference 18. The values given are the average for six compositions. We used $\kappa_3(0)$ not $\kappa_1(0)$ as was done by Ogasawara et al. [18].

[b]Reference 19. Average of two compositions Mo-15% Re and Mo-25% Re.

[c]Reference 20. This composition is very close to that used by Echarri et al. [4].

[d]Reference 21. The values given are the average of six compositions except for the values in the column headed Eq. (4) where only the three highest Ta compositions could be calculated.

[e]For this calculation the resistivity values were obtained from an earlier report by the same authors [24].

FIGURE CAPTIONS

Fig. 1 Normalized flux flow resistivity vs normalized applied transverse field for an annealed Nb-25% Ti wire 0.77 mm diam at T_B = 4.18 K (t = 0.38) where $H^*_{c2}(0)$ = 90.5 kG.

Fig. 2 The lower critical field vs temperature for three Nb-Ti alloys. The measurements were made on a bundle of six 2.6 cm lengths of wire each 0.77 mm in diameter. The values at T = 0 K determined by a smooth extrapolation of the data are listed in Table I.

Fig. 3 Magnetization vs applied axial field for a bundle of six 2.6 cm lengths of annealed Nb-5% Ti wire each 0.77 mm in diam at T_B = 7.2 K (t = 0.765).

Fig. 4 The measured first generalized Ginzburg-Landau kappa, $\kappa_1(T) = H_{c2}(T)/\sqrt{2}\, H_c(T)$, vs reduced temperature ($t = T_B/T_c$) for three Nb-Ti alloys compared with the values at T = 0 K obtained from Table II.

Fig. 5 The measured values of $H_{c1}(T)/H_c(T)$ vs reduced temperature ($t = T_B/T_c$) for the three Nb-Ti alloys compared with theory near T_c.

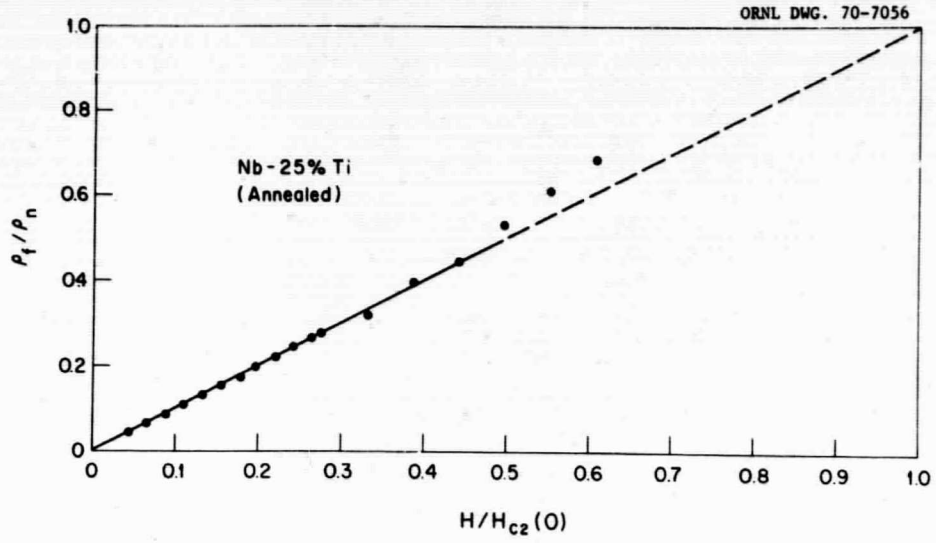
ORNL DWG. 70-7056
Nb - 25% Ti
(Annealed)
ρ_f/ρ_n
$H/H_{c2}(0)$
0
0.2
0.4
0.6
0.8
1.0
0.1
0.3
0.5
0.7
0.9

Fig. 1

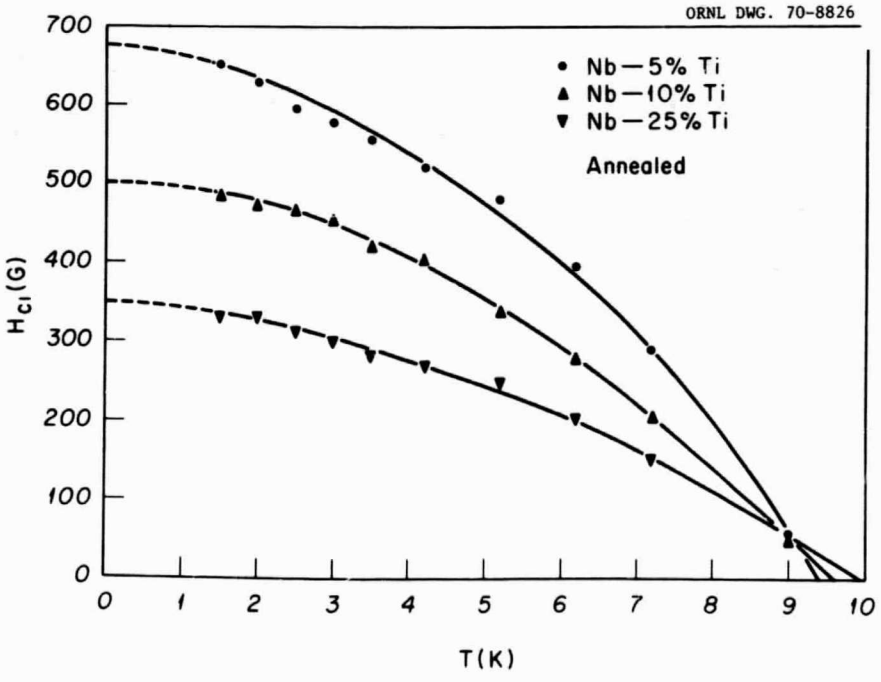
ORNL DWG. 70-8826
Nb—5% Ti
Nb—10% Ti
Nb—25% Ti
Annealed
$H_{c1}(G)$
T(K)
0
100
200
300
400
500
600
700
1
2
3
4
5
6
7
8
9
10

Fig. 2

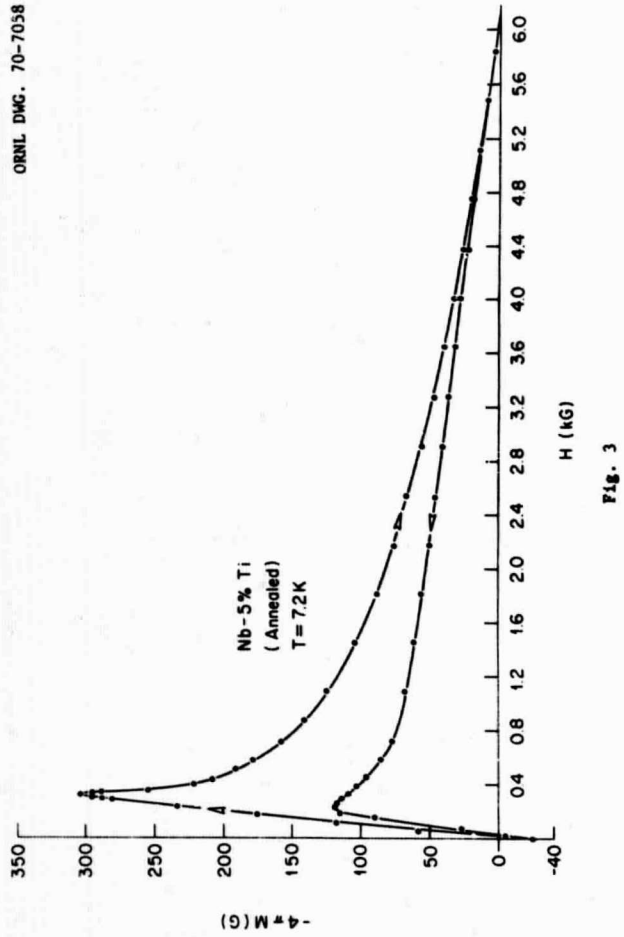
ORNL DWG. 70-7038
Nb-5% Ti
(Annealed)
T=7.2K
H (kG)
-4πM(G)
350
300
250
200
150
100
50
0
-40
04
0.8
1.2
1.6
2.0
2.4
2.8
3.2
3.6
4.0
4.4
4.8
5.2
5.6
6.0

Fig. 3

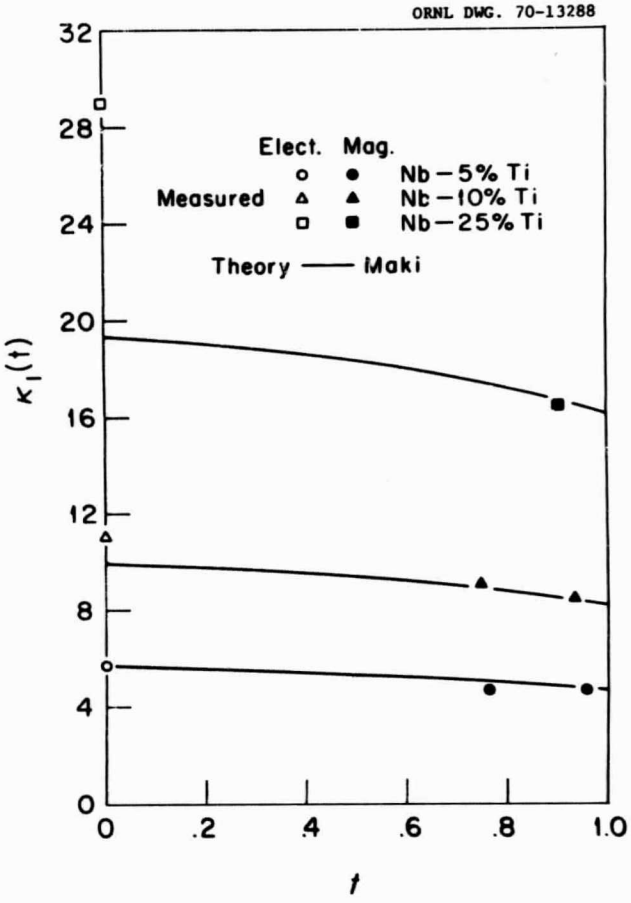
ORNL DWG. 70-13288
Elect. Mag.
Measured
Nb – 5% Ti
Nb – 10% Ti
Nb – 25% Ti
Theory —— Maki
$\kappa_1(t)$
t
0
4
8
12
16
20
24
28
32
0
.2
.4
.6
.8
1.0

Fig. 4

ORNL DWG. 70-13289

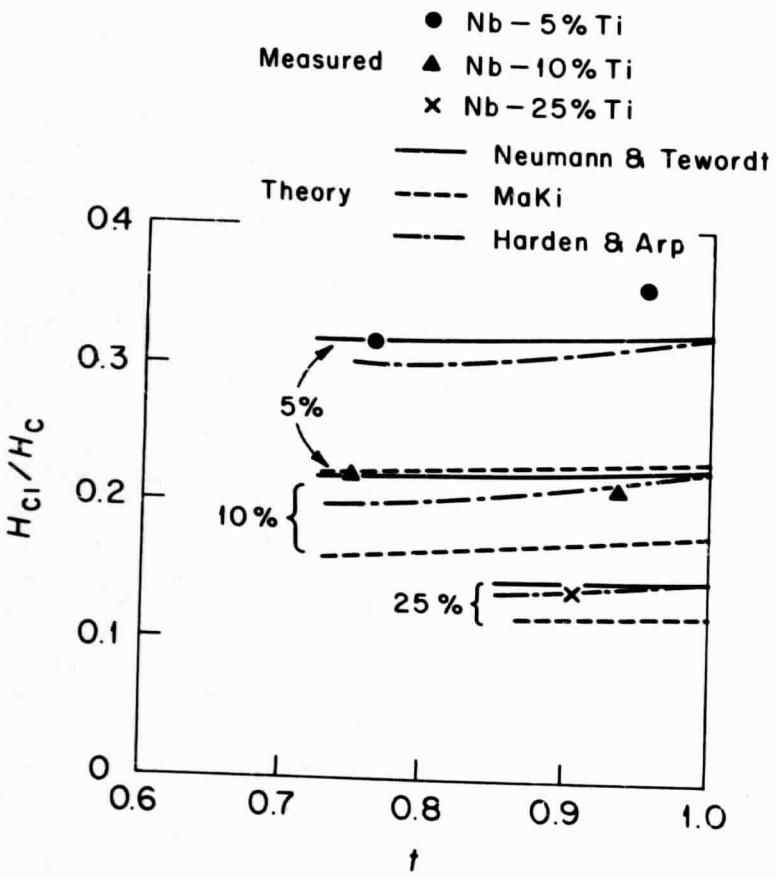

Fig. 5